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AUTHOR(S): P. W. Milonni
J. R. Ackerhalt
M. E. Goggin

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QUANTUM-MECHANICAL ASPECTS OF CLASSICALLY CHAOTIC DRIVEN SYSTEMS

P.W. Milonni, J.R. Ackerhalt, and M.E. Goggin
Theoretical Division (T-12)
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

Introduction

The onset of chaos plays an important role in classical models of multiple-photon excitation, dissociation, and ionization. In particular, the chaotic motion of trajectories in phase space can lead to a diffusive excitation in which the average energy of the driven system is proportional to the time. The question is whether any aspects of this stochastic excitation carry over into fully quantum-mechanical treatments.

Obviously this question is closely tied to that of "quantum chaos." Classically, chaos may be defined unambiguously by the existence of at least one positive Lyapunov exponent, implying the property of "very sensitive dependence on initial conditions." Classically chaotic systems have a continuous component in their power spectra, as opposed to regular, quasiperiodic motion in which power spectra consist of discrete, delta-function peaks. Classically chaotic systems also exhibit decaying correlations.

Quantum systems with discrete energy levels are obviously quasiperiodic, and therefore cannot exhibit chaos in the classical sense of a positive Lyapunov exponent. Here we are interested in driven quantum systems, and in particular in atoms and molecules in laser fields. For periodically driven quantum systems it has been established that (a) the dynamics is recurrent, [1] and (b) a quantum localization effect can suppress the classical diffusion in phase space and the consequent stochastic excitation. [2] Since the recurrence time can be very long and experimentally uninteresting, [3] we will focus our attention here on whether stochastic excitation is possible in a driven quantum system. [4]

The Kicked Pendulum

The kicked pendulum (rotor) is the system with Hamiltonian

$$H = p_\theta^2/m\ell^2 - (m\ell^2\omega_o^2)\cos\theta \sum_{n=-\infty}^{\infty} \delta(t/T - n) \qquad (1)$$

and equations of motion

$$\dot{p}_\theta = -(m\ell^2\omega_o^2)\sin\theta \sum_n \delta(t/T - n) \tag{2a}$$

$$\dot{\theta} = p_\theta/m\ell^2 \tag{2b}$$

m and ℓ are the pendulum mass and length, ω_o is the natural frequency for small displacements, and T is the period between kicks. For $T \to 0$ the force is on continuously and of course the system is integrable (not chaotic).

From (2) we obtain by integration the standard mapping

$$P_{n+1} = P_n + K\sin\theta_n \tag{3a}$$

$$\theta_{n+1} = \theta_n + P_{n+1} \tag{3b}$$

where $P_n = (T/m\ell^2)p_n$, $K = (\omega_o T)^2$, and p_n, θ_n are the values of p_θ, θ just before the nth kick. The advantage of studying such kicked systems, of course, is that the integration of trajectories is replaced by the iteration of a discrete map.

We are interested here in the energy gained by the kicked pendulum. For large K there is chaos and it is found that the energy, averaged over classical initial conditions, grows in a diffusive fashion:

$$\langle P^2 \rangle \approx \tfrac{1}{2}K^2 t \tag{4}$$

Such results were first discussed by Casati, et al. [5] These authors gave the following intuitive explanation for the role of chaos in the diffusive energy growth. From (3) it follows that

$$(P_n - P_o)^2 = K^2 \sum_{i=0}^{n-1} \sum_{j=0}^{n-1} \sin\theta_i \sin\theta_j \tag{5}$$

If the θ_i are treated as uniformly distributed random variables then the average of (5) over the θ_i is proportional to n. Thus we can see how chaos can give rise to a linear dependence of average energy on time (in this case n), i.e., stochastic excitation.

The question naturally arises as to how such results might carry over into quantum mechanics. It is found in numerical experiments for the quantum kicked pendulum that the energy expectation value at first increases and then shows a strong saturation behavior. [5] In other words, the stochastic excitation and phase-space diffusion predicted classically is strongly suppressed in the quantum dynamics.

This quantum suppression of the classical diffusive behavior was related by Grempel, et al. [2] to the Anderson localization of a particle in a one-dimensional lattice with random site energies. The basic idea of their elegant argument is that there is a mathematical correspondence between the kicked pendulum and the one-dimensional tight-binding model, such that the lattice points of the latter correspond to the integer values of quantized angular momentum in the former. The random diagonal terms of the tight-binding model correspond to a pseudorandom sequence

$$T_n = \tan[\tfrac{1}{2}(\omega - E_n/\hbar)T] = \tan[\tfrac{1}{2}(\omega T - n^2\tau)], \quad \tau = \hbar T/m\ell^2 \qquad (6)$$

in the kicked pendulum. That is, if τ is an irrational multiple of 4π then the sequence $\{T_n\}$ is effectively random, having decaying correlations and a broadband power spectrum. This pseudorandom sequence arises from the nature of the discrete energy spectrum of the (quantum) kicked pendulum, and shows that the Anderson-like localization in phase space is a quantum effect.

Thus at least some features of classical chaos must be mitigated by quantum mechanics. However, it should be noted that the assumption of periodic kicking is very important here. For quasiperiodic kicking, for instance, it is found that diffusive energy growth can occur over a very long time interval without any evidence of quantum suppression. [6,7] In this connection it has been found experimentally that Rydberg hydrogen atoms are less stable against ionization in two-frequency microwave fields than in single-frequency fields. [8]

Classical Models of Multiple-Photon Excitation and Dissociation

The stochastic excitation of the kicked pendulum results from resonance overlap [4,9,10] and the onset of widespread chaos. The concept of resonance overlap is especially useful for driven systems because the driving frequency can probe the various classical nonlinear resonances. Resonance overlap ideas have been applied in classical models of infrared multiple-photon dissociation by Jones and Percival

[11] and Galvao, et al. [12] We have also analyzed a classical model of multiple-photon excitation that is able to account for the experimental observation that the number of photons absorbed by a polyatomic molecule is determined primarily by the fluence of the laser pulse and not the intensity. [13] More recently we have shown that certain model-dependent features are removed when rotations are taken into account in this classical model. [14]

There are obvious questions. How reliable are the classical models? How, if at all, does the classical chaos manifest itself in a fully quantum-mechanical description of these driven systems? Is stochastic excitation possible in driven quantum systems? We do not yet have complete answers to these questions. We therefore turn our attention next to a simple but fairly realistic model of a molecule in a sinusoidal field, and address these questions for this specific model.

The Driven Morse Oscillator

We consider the system with Hamiltonian

$$H = p^2/2m + D(1 - e^{-\alpha x})^2 - dxE_L\cos\omega_L t \qquad (7)$$

D and α are the dissociation energy and range parameter, respectively, of the Morse potential, and d is the dipole gradient. Near the bottom of the well we have approximately harmonic motion with frequency $\omega_o = (2D\alpha^2/m)^{1/2}$. We write the equations of motion in the scaled form

$$d^2X/d\tau^2 = - (4/B^2)(e^{-X} - e^{-2X}) + 2K\cos\mu\tau \qquad (8)$$

where $\tau = (DB^2/\hbar)t$, $X = \alpha x$, $\mu = \hbar\omega_L/DB^2$, $K = dE_L/\alpha DB^2$, and the dimensionless parameter $B = (\hbar^2\alpha^2/2mD)^{1/2}$. We use parameters corresponding to the HF molecule, for which there are 24 bound states of the Morse potential. In these scaled variables the Schrödinger equation for the driven Morse oscillator is

$$i\partial\psi/\partial\tau = - \partial^2\psi/\partial X^2 + B^{-2}(1 - e^{-X})\psi - KX\cos(\mu\tau)\psi \qquad (9)$$

The first question concerns the comparison of the classical and quantum theories of the driven Morse oscillator. This question was addressed some time ago by Walker and Preston, [15] and their results

have often been invoked to justify classical models of multiple-photon excitation. Basically their conclusions were that the classical theory is fairly reliable for the scale and gross features of things like average energy absorbed, but that it is not very reliable near multiphoton resonances. We have recently extended this investigation to include the possibility of dissociation; our approach is to solve the partial differential equation (9) numerically without using a basis-state expansion for ψ. [16] Our results corroborate those of Walker and Preston, but also reveal that the classical and quantum predictions may differ substantially near higher-order classical nonlinear resonances.

To be more specific, let us note that for the Morse oscillator a classical resonance occurs when the laser frequency ω_L satisfies

$$\omega_L = N\omega(J) = N\omega_0\sqrt{1 - E/D} \tag{10}$$

where E is the oscillator energy, $\omega(J) = \omega_0(1 - \omega_0 J/2D)$, and N = 1,2,3... We find that, near such a classical resonance with $N > 1$, the classical theory typically predicts a considerably lower threshold field strength for dissociation than the quantum theory. Similarly near a quantum multiphoton resonance the classical theory typically predicts a considerably higher threshold than the quantum theory. Near a classical $N = 1$ resonance or a quantum single-photon resonance, the classical theory seems fairly reliable, and in fact the quantum threshold for dissociation may be predicted fairly accurately using Chirikov's resonance overlap criterion without taking higher-order resonances into account. [16]

The second question is whether classical chaos manifests itself in any way in the quantum theory. We prefer to phrase the question somewhat differently: how, if at all, does the classical resonance overlap manifest itself quantum mechanically? The reason for this preference is that the pre-dissociation dynamics is transient and it is not clear to us how to rigorously define "transient chaos." (The definition of the Lyapunov exponents involves the long-time system dynamics.) On the other hand it is clear in the classical dynamics that resonance overlap is necessary for the onset of dissociation, and we have already remarked that the dissociation threshold may be predicted fairly well using the resonance overlap criterion.

Now in the classical theory the width of each N-resonance turns out to be proportional to the square root of the electric field strength. This is a well-known consequence of the fact that a driven one-dimensional system may be approximated by the pendulum system near an isolated resonance. Based on semiclassical quantization ideas, then, we would be led to expect that the number of quantum levels that are strongly coupled by the applied field should be proportional to the square root of the electric field strength. That is, the number of quantum levels mixed by the field in the classical resonance overlap

regime, Δn, should be proportional to $\sqrt{E_L}$. We have found this prediction to hold up quite well in our numerical experiments with the driven Morse oscillator. We have also provided a heuristic explanation based on Rabi splittings of the unperturbed levels. [16]

Of course it should not be surprising that the number of levels mixed by the field should increase with the field strength. Based on this result, we perhaps have a qualitatitive understanding of how the classical resonance overlap translates into the quantum description. Specifically, the overlap of classical resonance zones corresponds to the mixing of quantum energy levels. Classically, resonance overlap results in diffusive motion in phase space and dissociation. Quantum mechanically, it is the spread of population with increasing field strength that gives rise to dissociation.

The pre-dissociation quantum dynamics is of course quasiperiodic. However, if many (discrete) levels are mixed by the field the dynamics can be quite complicated, perhaps enough so to justify statistical assumptions about the laser-molecule dynamics. For instance, the dynamics of a quasiperiodically kicked two-level atom is quasiperiodic, but it can nevertheless be ergodic on the Bloch sphere. [7] Furthermore the autocorrelation of the state vector can be regarded as a decaying function of time for practical purposes, with recurrences occurring only on long time scales. Such non-chaotic but "complicated" behavior is reminiscent of Slater's theory of unimolecular reactions, which invokes a large number of incommensurate frequencies to derive certain "statistical" features of quasiperiodic dynamics. [17]

In other words, quantum effects can certainly suppress the degree of chaotic behavior possible classically, but this does not rule out the possibility that the evolution of the state vector may proceed in a very complicated way, perhaps allowing for effectively statistical behavior that classically can be attributed to chaos. From this point of view it is not surprising that "stochastic" excitation is possible in quantum systems with quasiperiodic driving, [6] because a driving force made up of just a few incommensurate frequencies can act effectively as a "stochastic" force. [7] For such purposes quantum chaos in the rigorous sense may be impossible but also unnecessary.

Acknowledgements

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